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Abstract

The primary objective was to determine the aluminium (Al) content of selected foods and food products in the USA which contain Al as an approved food additive. Intake of Al from the labeled serving size of each food product was calculated. The samples were acid or base digested and analyzed for Al using electrothermal atomic absorption spectrometry. Quality control (QC) samples, with matrices matching the samples, were generated and used to verify the Al determinations. Food product Al content ranged from < 1 to 27,000 mg kg^{-1}. Cheese in a serving of frozen pizzas had up to 14 mg of Al, from basic sodium aluminium phosphate; whereas the same amount of cheese in a ready-to-eat restaurant pizza provided 0.03-0.09 mg. Many single serving packets of non-dairy creamer had ~ 50 to 600 mg Al kg^{-1} as sodium aluminosilicate, providing up to 1.5 mg Al per serving. Many single serving packets of salt also had sodium aluminosilicate as an additive, but the Al content was less than in single-serving non-dairy creamer packets. Acidic sodium aluminium phosphate was present in many food products, pancakes and waffles. Baking powder, some pancake/waffle mixes and frozen products, and ready-to-eat pancakes provided the most Al of the foods tested; up to 180 mg/serving. Many products provide a significant amount of Al compared to the typical intake of 3-12 mg/day reported from dietary Al studies conducted in many countries.

Introduction

Aluminium (Al) is distributed throughout the environment because of its presence as the third most abundant element on earth. Concern about Al toxicity to humans has persisted since the demonstration that it has the potential to be a neurotoxicant (Döllken 1898; Gies 1911; Anon. 1913; WHO 1997; Yokel and Golub 1997; ATSDR 1999). Aluminium can be toxic to bone, bone marrow and the nervous system. There is a proposed relationship between Al in the brain and the occurrence of Alzheimer's disease and other neurodegenerative disorders (Exley 2001). Exposure of humans to Al is mainly from food, water, airborne dust, antiperspirants, immunizations, allergy injections and antacids. Food is the single largest contributor of Al intake for the typical human (Yokel and McNamara 2001). Aluminium is present in food naturally, from its addition as food additives, and through contact with Al used in food preparation and storage (Pennington 1987). The U.S. Food and Drug Administration (FDA) permits the use of Al salts as generally recognized as safe (GRAS) food additives (21 CFR 182). Canada permits this practice, as described in Part B Foods Division 16 of its Food and Drug Regulations (http://www.hc-sc.gc.ca/food-aliment/friia-raaii/food_drugs-aliments_drogues/act-loi/e_index.html). The Canadian Food and Drugs Act regulations are similar to the FDA's. The fifteen member states of the European Union, as well as Norway and Iceland, allow the use of Al as a food additive (http://europa.eu.int/eur-lex/en/consleg/pdf/1995/en_1995L0002_do_001.pdf) (http://193.120.54.7/publications/reports/Legislation_Food_Additives.pdf), as does the United Kingdom (http://www.legislation.hmso.gov.uk/si/si1995/Uksi_19953187_en_5.htm#sdiv3) and

Australia and New Zealand (http://www.foodstandards.gov.au/mediareleasespublications/publications/shoppersguide/foodadditivesnumeric1680.cfm). However, Poland apparently did not permit Al-based food additives (Starska 1993). Examples of permitted Al additives include basic sodium aluminium phosphate (SALP) (E 554 in the EC) as an emulsifying agent in pasteurized process cheese, cheese food and cheese spread (21 CFR 133); acidic SALP (E 541 in the EC) as a leavening agent in cereal foods and related products (21 CFR 137); and sodium aluminosilicate (SAS) (E 554 in the EC) as an anti-caking agent. There have been many reports of the Al content of foods and beverages, including a summary of reports from many countries up to the mid-1980s (Pennington 1987), a study in the UK from 1985 to 1993 (UK MAFF 1993) and another in 1997 (Ysart *et al.* 2000) and a study of foods in Germany from 1988 and 1991 (Müller *et al.* 1998). These studies identified approved food additives as the primary source of Al in food.

The annual use of Al-containing food additives in the USA has probably increased over the past several years due to the many recent additions of SALP to foods. In 1982, four million pounds of Al were used as food additives in the USA (NRC 1984). There are two major companies in the global phosphates industry, Rhodia and Astaris. Rhodia completed an expansion in 1998 to its Nashville plant to increase its SALP capacity by 15% (1998) and Astaris was investing in capacity additions in 2001 because demand for its SALP exceeded capacity (Challener 2000).

Few investigations have been conducted recently in the USA to determine the Al content of food products that contain Al as an approved additive. Therefore we conducted a study to document the current levels of Al in selected foods and food products. The primary aim of this study was to determine the Al content of representative food products available commercially in the USA. To achieve this aim, methods were developed and validated to prepare and analyze Al in the food and food products. The analysis of Al is challenging because of its low concentrations in some foods and the potential for contamination during sample preparation and analysis (Vinas *et al.* 2001). To validate the procedures we developed, QC samples were prepared because reference or certified materials that could be used as QC standards were not available commercially in relevant matrices, e.g., Al in food products similar to those we tested. Food products selected for study were those containing Al as well as similar products which did not list Al as an ingredient. The representative food products studied included pizza cheese, pizza crust, cheese, grains, baked goods, cake mixes, baking powder, pancakes and waffles, non-dairy creamers, salt, and pickle relish. The amount of Al in a serving of each of these representative food products was calculated and compared to the reported values of typical human daily Al intake from the diet.

Material and Methods

Approximately ninety-five single sample purchases were made locally during 2003-2004, unless otherwise noted, of different food products in selected categories, as shown in the tables. The products purchased are nationally available and house brands which are presumed to be available nationally. They were selected to be representative of products that did, or did not, have added Al, according to the Nutrition Facts label. The purchase locations were varied, but since all the products were national brands or house brands of national chains, it was assumed the products would be the same if purchased elsewhere. Some were known to contain Al from their list of ingredients. Most were prepared for electrothermal atomic absorption spectrometric (ETAAS) analysis of Al, as described (Yokel and Melograna 1983). Many of the samples analyzed were powder-like materials, assumed to be homogeneous. The relish samples were homogenized in a Waring® blender before analyzing the samples. A ~ 35 mg sample of the food product, as purchased, was accurately weighed in a pre-weighed Teflon® screw cap container. All the pizzas studied had cheese. A portion of pizza (averaging 13 gm) was weighed and the cheese carefully separated from the remaining ingredients and weighed to determine the average weight of the cheese as a percentage of the sample. This process was repeated for 2 or 3 additional replicate samples. Non-cheese samples were digested using one ml of 70:30 HNO_3:H_2O_2 in closed Teflon® containers overnight on a 100 °C hot plate (temperature determined by thermocouple), followed by acid and liquid evaporation typically at 90 °C for 5 to 6 hours and then at 135 °C for 2 to 3 hours. After acid evaporation the sample residue was dissolved by addition of 0.14 ml of 70% HNO_3 in the Teflon® container on a hot plate at ~ 100 °C overnight. When necessary to dissolve the residue, an additional 0.21 ml of 70% HNO_3 was added. The resulting solution was then diluted with H_2O and Mg addition to produce a final matrix of 0.2% HNO_3 and 2.5 mM Mg. Samples were further diluted in this matrix, when necessary, to generate an Al concentration within the standard curves used for ETAAS, using a Perkin Elmer 4100 ZL spectrophotometer. Standards were prepared from an atomic absorption standard solution (Aldrich Chemical Company, Milwaukee, WI, USA) in the same matrix. Samples containing SAS required addition of 48% HF to solubilize the Al.

Another method was developed for cheese samples because they did not completely dissolve in 70:30 HNO_3:H_2O_2 owing to their high fat content, potentially resulting in poor Al analysis. Furthermore, digestion of cheese using HNO_3 has been stated to be hazardous due to its high fat content by (Delves *et al.* 1989) who developed a method to remove the lipid fraction by solvent extraction with petroleum ether and then back extraction of the Al into 0.1 M HNO_3. We tried to apply this method with back extraction into 0.1 M HNO_3 and alternatively 5 M KOH. This did not work because the high volatility of petroleum ether made quantification of Al in it difficult using ETAAS. Therefore, we could not verify recovery of Al, by back extraction, from lipid. Recovery of Al from the organic phase was deemed essential because full recovery was not obtained from the aqueous phase. We observed that the KOH dissolved the cheese.

Therefore we developed a new method to digest cheese to create homogenous samples for Al analysis. A sample of ~ 30 to 35 mg of cheese was digested in 1 ml of 1 % KOH overnight on a hot plate at ~ 100 °C. This resulted in a solution with some lipid particles floating on the surface. This was sonicated using a Fischer Scientific Sonic Dismembrator Model 500 for 20 seconds, with a 5 second pause after the first 10 seconds, at 22 % amplitude, producing a cloudy solution with no floating particles. The samples were then diluted immediately, into a final matrix of 0.001 % KOH/ 2.5 mM Mg and analyzed for Al using by ETAAS. The pH of 0.001 % KOH is ~ 7.5, acceptable for injection into the graphite tube for ETAAS. Standards were prepared from an Al atomic absorption standard, as noted above, in the same matrix.

Blanks were included with each batch of samples prepared for Al analysis to estimate the background Al contamination of the procedure. Blanks were Teflon® containers that did not contain sample but received the same treatments as the samples.

An Al recovery study of this method of cheese digestion was conducted by adding 37.9 mg Al kg^{-1} to ~ 35 mg samples of Swiss, American and cheddar cheese and processing the samples for Al analysis.

The recommended conditions for Al analysis by ETAAS were modified, as shown below, and utilized for analysis of all samples. An inert gas (argon) flow rate of 250 ml/min was used. Twenty µl injections of samples and standards were made.

Step	Temperature (°C)	Ramp time (s)	Hold time (s)
1 (Dry)	110	5	20
2 (Dry)	130	5	45
3 (Char)	1600	10	20
4 (Atomization)	2400	0	3
5 (Clean)	2500	1	1

Different QC samples were prepared for each type of food analyzed. All food samples were initially analyzed at least once to ascertain their approximate Al concentration. Quality control samples were then created by addition of sufficient Al to a food containing very little Al to bring the total Al into the range of typical samples of that type of food. For example, Al was added to sample D25, which had ~ 11 mg Al kg^{-1} (table 4) to bring the Al concentration to ~ 580 mg kg^{-1} (table 8), and into the range of Al concentrations seen in pancake, hot cake and waffle products containing considerable Al (table 4). To create the QC samples AlK_2SO_4 or Kasal® (70% basic SALP and 30% dibasic sodium phosphate) was added to the solid samples and an atomic absorption standard solution of Al was added to the homogenized semi-liquid sample. The added Al was introduced into a small amount of the food. This was diluted with further addition of food to try to achieve uniform Al distribution throughout the QC sample. Cheese QC samples were prepared by adding Kasal® to a 9:1 cheese:water mixture and microwaving it for 13 and 5 second cycles, separated by 15 seconds during which time the mixture was stirred. Quality control samples containing the solid food products were produced by adding Al and mixing them well for approximately 20 minutes in a Waring® blender. A sample of relish was homogenized in the blender, Al was added, and the mixture vortexed for 10 to 15 minutes to make that QC sample. Five replicate aliquots of each QC were prepared for Al analysis, using the procedures described above for samples, and analyzed. The QC samples were then included with at least one batch of every sample analyzed to validate the sample preparation and analysis procedure. Results were accepted when the Al concentration of the QC samples was within 10% of the mean of the five replicate aliquots. The cheese QC was included with analysis of samples containing pizza cheese and other cheese products. Similarly non-dairy creamer, salt, baking mix and relish QCs were included with their respective samples. Cake mix QC was included with the cake mix samples and hot cocoa mix. Waffle QC was included with pancake, hot cake and waffle samples.

The Al concentration in samples was determined in three or four independently conducted analyses of a single purchase of the food product. The one exception is B05 in table 2, for which 2 separate purchases were made from different locations. The Al concentration results are reported as the mean ± S.D. of three replicate observations, and for samples analyzed four times, as indicated by * in the tables. The results are based on the weight of the product as purchased.

Results

The recovery of Al from the Swiss, American and cheddar cheeses was within 10% of complete, suggesting the new method to digest cheese was acceptable. The method was then used to digest all cheese samples.

The relative standard deviation of the five replicates of each QC sample was < 10%, indicating that these samples were sufficiently homogenous and the analysis consistent enough to use these as QC samples to validate the methods of samples subsequently analyzed.

The results of Al determination in the food products are shown in tables 1 to 7. The Al contributing ingredients, according to the product's Nutrition Facts label, were SALP or SAS. The Al concentration determined in the samples was converted to Al in a labeled recommended serving to enable comparisons among products and to reported daily Al intakes.

The labeled recommended serving size of the frozen pizza contained an average of 14 g of Al in the cheese (table 1). The Al in the cheese of a serving of ready-to-eat restaurant pizza and other cheese products which do not mention use of Al as an additive was ~ 0.04 to 0.7 mg (tables 2 & 3). The results in table 1 show that Al is an ingredient in many frozen pizzas, and when present provides a considerable amount of Al. Processed cheese slices do not commonly contain added Al. When Al is present, less is present than in most frozen pizza cheeses (tables 1 & 3). In contrast to frozen pizzas, the cheese in ready to eat (restaurant, carry-out, take-away) pizzas (table 2) had very little Al, suggesting Al had not been added. As the serving sizes of ready-to-eat pizzas are not provided with the product, we used the average amount of cheese and crust of frozen pizzas to estimate the serving size of cheese in ready-to-eat pizzas (table 2) to enable calculation of Al consumption from a typical serving. One pizza crust contained a large amount of Al, whereas Al did not appear to be an additive in others (tables 1 & 2).

(tables 1, 2 & 3 about here)

Food products such as flour and cake mixes and foods such as cinnamon rolls, pancakes and waffles had variable Al content (table 4). Some products had little Al whereas some similar products had considerable amounts. The baking powders tested contained considerable Al (table 4).

(table 4 about here)

It appears that SAS is added to most non-dairy creamer products as an anti-caking agent (table 5). One recommended serving size provides 0.1 to 1.5 mg Al. SAS is also routinely added to single use packets of salt (table 6). In contrast, only some salt sold in multi-use containers had added Al, according to the list of ingredients and the results (table 6). Al intake from one serving of Al-containing salt was ~ 0.1 to 0.2 mg whereas the samples without added Al provide negligible Al. Pickle relish often contains Al (table 7).

(tables 5, 6 and 7 about here)

The QC samples prepared by adding a known amount of Al to samples that contained little Al are shown in table 8. The brand codes in table 8 correspond to the same brands in the previous tables, which report the initial Al concentration of these food products.

Discussion

There are many reports of the Al content of various components of the human diet. The more complete compilations include (Pennington 1987; Schenk *et al.* 1989; UK MAFF 1993; Müller *et al.* 1998; ATSDR 1999). There are numerous reports of Al in milk and infant formulae. For example, the Al concentrations in cow- and soy-milk-based infant formula in the UK ranged from 0.03 to 0.2 and 0.53 to 1.34 mg L^{-1}, respectively (Baxter *et al.* 1990; Baxter *et al.* 1991). The Al concentrations in 44 brands of milk powders and simulated milk powders in Singapore were generally < 1 mg, although some ranged up to 15 mg kg^{-1} powder in a soy formula (Bloodworth *et al.* 1991). In 282 cans of infant formulae and evaporated milks sold in Canada, evaporated milk and milk-based formulae generally had lower Al concentrations, 0.022 to 0.34 and 0.01 to 2.49 mg kg^{-1}, respectively, than soy-based formulae (0.40 to 19 mg kg^{-1}) (Dabeka and McKenzie 1990). Soy-based formulae had the highest Al concentration among the infant formulae tested recently in Spain, 0.313 to 3.48, vs. 0.068 to 2.72 mg L^{-1} for other types of formulae (Navarro-Blasco and Alvarez-Galindo 2003). Soy, cow, and breast milk were found to have quite low Al concentrations, 0.005 to 0.285, 0.004 to 0.033 and 0.003 to 0.079 mg L^{-1}, respectively (Baxter *et al.* 1991). Therefore, infants consuming soy-based formulae receive much more Al than those consuming milk-based formulae.

Tea contributes a variable percentage of Al to daily intake. The many reports of the Al concentration in brewed tea suggest the average is ~ 3 to 4 mg L^{-1}; 10-fold or more higher than in coffee (Pennington 1987; Pennington and Jones 1989; Schenk *et al.* 1989; Sherlock 1989; Baxter *et al.* 1990; Müller *et al.* 1998; Sepe *et al.* 2001). Soft drinks and fruit juices generally contain < 1 mg Al l^{-1} (Pennington 1987; Pennington and Jones

1989; Baxter *et al.* 1990; Müller *et al.* 1998; Sepe *et al.* 2001). In countries where Al from other sources is relatively small and tea consumption relatively large, as in the UK, tea has been estimated to contribute ~ 50% of the total daily Al intake (UK MAFF 1993).

There are many studies reporting the Al content of unprocessed and processed foods (Greger 1985; Pennington 1987; Sullivan *et al.*, 1987; Sherlock 1989; Müller *et al.*, 1998). However, there are no recent studies focusing on processed food products in the USA. This study determined the Al concentration in many food and food products that typically contain Al as a food additive. The results represent the current concentration of Al in many USA food products; both those containing Al as a food additive and similar products that do not declare this. The Al concentrations reported in this study are the sum of those inherently present, contributed by food additives and that introduced by processing and storage because the methods used do not distinguish among these sources. Similarly, the addition of ingredients to mixes, such as cake or pancake mixes, could increase the Al content due to the additions or the processing and storage of the final food product.

Frozen pizza that listed SALP as an additive generally had ~ 200 to 750 mg Al kg^{-1} cheese (table 1). The cheese from frozen pizza products that did not list Al as an additive and ready-to-eat pizza had only a few mg Al kg^{-1}, similar to a natural cheese (tables 1, 2 & 3). These results are similar to previous reports from several countries of a few mg Al kg^{-1} in natural cheese (Pennington 1987) and from the USA (Greger 1985; Schenk *et al.* 1989), the UK (Delves *et al.* 1989), Italy (Favretto 1990), India (Rao and Rao 1993), and Germany (Müller *et al.* 1998). Our findings are also similar to the several hundred up to 750 mg Al kg^{-1} in processed cheese in the United States (Gormican 1970; Greger 1985;

Pennington and Schoen 1995). Thus the Al content of natural cheese and processed cheese containing added Al has been reasonably constant for a few decades.

This is the first report to compare the Al concentration in frozen prepared and ready-to-eat pizza products. The crust (grain product/bread) of frozen pizza that did not list Al as a food additive and in ready-to-eat pizza contained ~ 12 mg Al kg^{-1} (tables 1 and 2), whereas a frozen pizza listing Al as an additive (SALP) in the crust had ~ 200 mg Al kg^{-1} (table 1). An extensive review of the literature failed to reveal any previous reports of the Al concentration in pizza crust. The high concentration of Al in the crust of the pizza containing Al as an additive is consistent with baked goods representing the largest source of Al in the typical diet (Pennington 1987).

The Al concentration in some foods (baked goods) that did not list Al as an additive was much lower than products that listed, and many products that did not list, Al as an additive (table 4). It appears that Al is an additive in some products which do not declare this in the ingredient list. Our observations are consistent with many previous reports showing considerable amounts of added Al in grain-based foods, particularly those containing self-rising flour (Pennington 1987; Jorhem and Haegglund 1992; Müller *et al.* 1998; Ysart *et al.* 2000). The very high Al concentration in baking powder is also consistent with previous reports of 20,000 to 34,000 mg Al kg^{-1} (Holak 1970, 1972; Rajwanshi *et al.* 1997). However, there are reports of baking powders containing much lower Al concentrations (0 to 24 mg kg^{-1}) (Holak 1970, 1972; Schenk *et al.* 1989; UK MAFF 1993), suggesting Al is not essential in these products. We found considerable Al in many pancake and waffle mixes, frozen and ready to eat products; up to 1200, 600 and 1200 mg kg^{-1}, respectively (table 4). Previous reports found 5 to 9 mg Al kg^{-1} in pancake wheat in Germany (Müller *et al.* 1998), 11 to 100 mg Al kg^{-1} in pancake mixes (Schenk *et al.* 1989; UK MAFF 1993) and 2 to 69 mg Al kg^{-1} in pancakes (Varo *et al.* 1980b; Pennington and Jones 1989; Pennington and Schoen 1995; Dolan and Capar 2002). It appears that the use of Al in pancake mixes and prepared pancakes in the USA has increased compared to previous surveys.

We found salts that listed SAS as an ingredient contained ~ 125 to 200 mg Al kg^{-1} and a few salts that did not declare SAS to contain around 3 mg Al kg^{-1} (table 6). Salt was previously reported to have < 1 to 165 mg Al kg^{-1} (Varo *et al.* 1980a; Greger *et al.* 1985; Schenk *et al.* 1989; Müller *et al.* 1998).

Single-use packets of non-dairy creamer had ~ 110 to 600 mg Al kg^{-1}, whereas multiple-serving products contained ≤ 50 mg Al kg^{-1}. There are a few previous reports of Al in non-dairy creamers, which contained 25 to 140 mg Al kg^{-1} (Pennington and Jones 1989; Schenk *et al.* 1989), consistent with products currently available in the USA. Our findings of 40 to 73 mg Al kg^{-1} in pickle relish are consistent with a single report of 39 mg Al kg^{-1} in pickles with additives (Greger *et al.* 1985). The remaining previous reports of the Al concentration in pickles and a single report of pickle relish reported Al concentrations of < 1 to 13 mg kg^{-1}, below the products we tested (Pennington and Jones 1989; Schenk *et al.* 1989; Gramiccioni *et al.* 1996; Takeda *et al.* 1998).

A recent report of Al in convenience and fast foods noted the increased popularity of these foods (Lopez *et al.* 2002). Studying beef-, chicken-, fish-, pork- and egg-based foods and sauces in Spain, they found up to 4 mg Al/food portion for some chicken-based foods. The results of the present study show considerable Al in many convenience foods.

Most estimates of Al intake from 1970 to the present estimated average daily dietary Al intake to be ~ 6 mg (Sherlock 1989; WHO 1997). For example, daily dietary Al intake in the USA was estimated to average 8 to 11.5 mg for adults and 14 to 16 year old males (Pennington and Schoen 1995). It was estimated the Dutch consume an average of 3.1 mg Al daily (Ellen *et al.* 1990), those in the UK 3.4 mg daily (Ysart *et al.* 2000) and Italians ~ 5 mg Al daily (Gramiccioni *et al.* 1996). However, the individual range of daily Al intake is great. For example, in a early study, an average daily Al intake of 24.6 mg was obtained from a range of 3.8 to 51.6 mg for total diet composites calculated to provide 4200 cal/day (Zook and Lehman, 1965). Similarly, the estimate of 3.1 mg Al consumed daily by the Dutch was based on a range of 0.6 to 33 mg for different study subjects (Ellen *et al.* 1990). Many of the products tested in the present study could easily provide more Al in one serving than consumed in the average daily diet, and some products are often consumed more than once daily, leading to the potential for even higher daily Al intake. Examples of products containing considerable Al are frozen pizza (from the cheese, and sometimes the crust as well) and many foods (baked goods), particularly pancakes. Similar products, for example ready-to-eat pizzas and alternative brands of baked goods, which do not list Al as an additive, often had much less Al, enabling the consumer to reduce Al intake through product selection. The absence of Al in comparable products raises the question whether addition of Al is necessary in most cases, and suggests that it is not, creating the possibility that the addition of Al to food products could often be avoided, reducing Al exposure through foods. The data from this study clearly indicate several magnitude higher Al intake from the food products that have Al as an additive than those which do not.

The potential for Al to contribute to Alzheimer's disease (AD) is controversial. There have been many epidemiological studies designed to assess if there is a link between elevated Al in drinking water and the development of AD. Some have shown a small but statistically-significant increased odds ratio for dementia in humans who lived in areas with higher drinking water Al concentrations whereas some studies did not show an association (Rondeau and Commenges 2001). Al consumption from foods is typically > 10-fold greater than from drinking water. We have found, using ^{26}Al-labeled biscuit and cheese, that oral Al bioavailability from these representative foods is ~ 0.02 and 0.05% in the rat (Yokel *et al.* 2005). This is lower than previous estimates of 0.09% when Al was added to the diet as Al lactate in fruit juice (Greger and Baier 1983) and estimates of ~ 0.1% based on average daily urinary Al excretion compared to average daily Al intake from food (Powell and Thompson 1993; Priest 1993; Nieboer *et al.* 1995). The oral bioavailability of Al from drinking water has been estimated to be ~ 0.2 to 0.3% (Priest *et al.* 1998; Stauber *et al.* 1999; Yokel *et al.* 2001). Therefore food presents more absorbed Al than drinking water for the typical human.

Al can be mobilized from Al cookware, particularly by acidic and basic foods. For example, cooking tomatoes (pH 4.4) and rhubarb (pH 3.4) in an Al saucepan increased the Al content of the foods from ~ 0.16 µg Al/g wet weight to 21.5 and 42 µg Al/g, whereas cooking them in a Teflon-coated saucepan raised the Al concentrations to 0.30 and 0.37 µg/g (Fairweather-Tait *et al.*, 1987). Similarly, (Gramiccioni *et al.*, 1996) showed that preparing tomato sauce, French beans, pickles, coffee and tap water in Al cookware increased the Al content considerably, compared to glassware or stainless steel cookware. Food storage and processing are generally not major contributors to Al in food

(Pennington 1987; Sherlock 1989). However, when patients with chronic renal insufficiency who used Al kitchen utensils for > 1 year were divided into one group that continued to do so for 3 months and another group that used stainless steel utensils, the latter group showed a significantly greater decrease in serum Al and daily urine Al excretion. These results suggest Al kitchen utensils may be a significant Al source for this population (Lin *et al.* 1997). When foods provide more Al in the diet than cookware, as is usually the case, one might expect foods that contain considerable Al to have an even greater contribution to the Al body burden of patients with chronic renal insufficiency than produced by Al kitchen utensils. Contrary to this notion, it has been noted that no public health threat due to Al in food has been detected (Humphreys and Bolger 1997) and that the risk of adverse effects of dietary Al, if there are any, is extremely low (Soni *et al.* 2001).

A preliminary study of 23 newly-diagnosed AD patients and 23 matched non-demented controls to ascertain the relationship between consumption of foods generally high in Al during the previous 5 years and dementia showed increased odds ratios for many food categories. However, the results were only significant for the category containing pancakes, waffles, biscuits, muffins, cornbread and corn tortillas. The odds ratio was not elevated for tea consumption, which the authors estimated might contribute ~ 25% of the daily Al intake in this population (Rogers and Simon 1999). In another study, oral Al absorption from an Al citrate drink was greater in 68 to 76 year old subjects diagnosed with AD than in age-matched controls, although there was not a significant difference between older patients and age-matched controls (Taylor *et al.* 1992). These small-scale studies suggest, but certainly do not provide strong evidence for, an increased risk of AD associated with Al in the diet and its absorption. Given that Al has no health benefits, but has the potential to produce toxicity, at least in people with impaired or absent renal function, some people may wish to avoid Al when it is practical to do so.

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table 1. Aluminium in frozen pizza cheese and crust.

Brand [a]	Pizza type	Al contributing ingredient [b]	Al concentration (mg kg $^{-1}$)	Serving size (g)	Approximate weight of the cheese or crust in one serving (g) [c]	Al / serving (mg)
Cheese						
A01	Extra cheese	NA	2.5 ± 1.1*	145	20.3	0.05
A01	Pepperoni	SALP	490 ± 260*	193	12.3	6.0
A01	Sausage, pepperoni, green peppers & onion – yellow colour cheese	SALP	650 ± 240*	204	14.8	9.7
A01	Sausage, pepperoni & green peppers & onion – white colour cheese	SALP	410 ± 150	204	6.2	2.6
A02	Italian cheese	NA	0.54 ± 0.38	131	15.1	0.008
A02	Italian sausage and pepperoni pizza	SALP	190 ± 60	198	9.2	1.8
A02	Sausage, Canadian style bacon and pepperoni	SALP	590 ± 270	204	9.2	5.5
A02	Pepperoni, peppers and onion	SALP	590 ± 140*	204	12.4	7.4
A03	Cheese	SALP	320 ± 150	121	21.5	6.9

A04	Original crisp crust	NA	3.0 ± 0.5*	139	14.7	0.044
A04	Sliced pepperoni - cheese	SALP	480 ± 180	145	16.1	7.8
A04	Sausage and pepperoni	SALP	750 ± 410	152	18.3	13.8
A04	Sausage & pepperoni with green peppers & onion – white colour cheese	SALP	680 ± 270*	155	3.1	2.1
A04	Sausage & pepperoni with green peppers & onion – yellow colour cheese	SALP	420 ± 140*	155	6.9	2.9
A05	Self rising crust four cheese	NA	1.7 ± 0.1*	139	16.6	0.028
Crust						
A06	Cheese stuffed	SALP	200 ± 30	120	80	16
A07	Original crust	NA	12 ± 9*	140	80	0.96

[a] Brand codes assigned to maintain anonymity.
[b] NA = Aluminium not listed as ingredient
SALP = Sodium aluminium phosphate
[c] Serving size as stated on the Nutrition Facts label.
[*] Values based on 4 replicate observations.

table 2. Aluminium in ready-to-eat restaurant (carry-out, take-away) pizza cheese and crust.

Brand	Pizza type	Al concentration (mg kg $^{-1}$)	Approximate weight of cheese or crust in a serving size (g) [a]	Al / serving (mg)
Cheese				
B01	Green pepper	2.6 ± 1.6	13.5	0.035
B02	Veggie	1.8 ± 0.2	13.5	0.024
B03	Olive	6.9 ± 1.7	13.5	0.093
B04	Green pepper	3.4 ± 0.7	13.5	0.046
B04	Extra cheese	3.1 ± 0.2	13.5	0.042
B05 – 2 purchases	Cheese	2.9 ± 0.4 4.5 ± 1.9	13.5	0.050
B06	Pepperoni and black olives	2.5 ± 0.9	13.5	0.034
Crust				
B07	Cheese	12 ± 7*	80	1
B08	Cheese [b]	12 ± 11	80	1

[a] Approximate weights are based on the average weights of the cheese and crust in Table 1, 13.5 and 80 g, respectively.
[b] Pizza purchased in Indiana.
* Value based on 4 replicate observations.

table 3. Aluminium in cheese

Brand	**Product**	**Al contributing ingredient [a]**	**Al concentration (mg kg $^{-1}$)**	**Weight of the cheese in one serving (g)**	**Al / serving (mg)**
C01	Cheddar cheese, sharp	NA	3.9 ± 3.9	28.4	0.11
C02	Processed American cheese slices	SALP	470 ± 200*	19	8.9
C03	Processed American cheese slices	SALP	14 ± 6*	19	0.27
C04	American pasteurized prepared cheese	NA	6.6 ± 4.4*	19	0.13
C05	Processed American cheese slices	NA	40 ± 55*	19	0.76

[a] NA = Aluminium not listed as ingredient
SALP = Sodium aluminium phosphate
* Values based on 4 replicate observations.

table 4. Aluminium in baking supplies, baked goods, pancakes and waffles.

Brand	Product	Al contributing ingredient[a]	Al concentration (mg kg $^{-1}$)	Serving Size (g)	Al / serving (mg)
D01	All-purpose enriched bleached pre-sifted flour	NA	19 ± 10	30	0.57
D02	Baking mix original	NA	830 ± 480	40	33
D03	Baking (biscuit) mix	NA	20 ± 5	32	0.64
D04	White corn meal mix self rising	NA	170 ± 120	34	5.6
D05	Self-rising white corn meal mix	NA	25 ± 10	25	0.62
D06	Baking powder	SALP	18000 ± 9000	0.6	11
D07	Baking powder	SALP	28000 ± 1000	1.1	30
D08	Carrot cake mix	SALP	440 ± 60	51	22
D09	Golden cake mix	NA	20 ± 2	52.4	1.2
D10	Angel food cake mix	NA	2 ± 2	37.8	0.09
D11	White cake mix	SALP	820 ± 150	43	35
D12	Pancake, waffle mix	SALP	1200 ± 600*	47	57
D13	Pancake, biscuit mix	NA	1080 ± 210	40	43
D14	Complete pancake mix	NA	670 ± 100	47	31
D15	Whole wheat buttermilk pancake mix	NA	19 ± 1	40	0.77

D16	Pancake mix	SALP	620 ± 460	43	27
D17	Buttermilk pancake mix	NA	70 ± 55	43	2.9
D17	Blueberry muffin mix bakery style	SALP	470 ± 210	56	26
D18	Blueberry muffin mix	SALP	490 ± 30	36	18
D19	Chocolate chip cookie refrigerated dough	NA	8 ± 5	25.5	0.22
D20	Chocolate chunk cookies refrigerated dough	NA	90 ± 61	25.5	2.3
D21	Buttermilk biscuits refrigerated dough	NA	30 ± 8	63.5	2
D22	Buttermilk scratch biscuits refrigerated dough	NA	98 ± 51	31	3
D23	Frozen Cinnamon Rolls	SALP	390 ± 220	44	17
D24	Frozen Cinnamon Rolls	NA	45 ± 18	44	2.1
D25	Frozen pre-baked waffle, buttermilk	NA	11 ± 8*	69.6	0.79
D26	Frozen buttermilk waffle	NA	600 ± 120	69.8	41
D27	Frozen buttermilk pancake	NA	300 ± 25	116	35
D28	Frozen raspberry pie	NA	3 ± 4	131	0.39
D29	Restaurant pancake [b]	NA	1200 ± 590	141	165
D30	Restaurant pancake [c]	NA	880 ± 100	141 (based on D29)	124
D31	Restaurant pancake	NA	450 ± 130	210	95

D32	Restaurant pancake	NA	530 ± 80	270	142
D33	Restaurant pancake	NA	560 ± 130	324	182
D34	Restaurant pancake	NA	430 ± 50	120	52
D35	Restaurant waffle	NA	43 ± 14	140	6
D36	Doughnut	NA	9 ± 6	60	0.6
D36	Raspberry filled doughnut	NA	7 ± 4	60	0.4
D36	Glazed cruller	NA	21 ± 7	60	1.3

[a] NA = Aluminium not listed as ingredient
SALP = Sodium aluminium phosphate
[b] Product purchased in Canada
[c] Product purchased in Utah.
[*] Values based on 4 replicate observations.

table 5. Aluminium in non-dairy creamer powder and cocoa

Brand	Type	Al contributing ingredient [a]	Al concentration (mg kg $^{-1}$)	Serving Size (g)	Al / serving (mg)
Single-serving non-dairy creamer packet					
E01		SAS	160 ± 60	2.7	0.44
E02		SAS	250 ± 100	2.7	0.68
E03		SAS	110 ± 40	2.4	0.26
E04		SAS	110 ± 50	2.4	0.26
E05		SAS	255 ± 40	2.1	0.54
E06		SAS	590 ± 140	2.5	1.5
E07		SAS	180 ± 40	2.5	0.45
Multiple-serving non-dairy creamer container					
E08	Creamer original	NA	7 ± 4	2	0.10
E09	Hazelnut flavour	SAS	50 ± 11	12	0.60
Cocoa and milk chocolate mixes					
E10	Hot cocoa mix	SAS	150 ± 80	20	3
E11	Hot cocoa mix with mini marshmallows	NA	25 ± 8	28.3	0.67
E12	Hot cocoa mix with milk chocolate marshmallows	NA	6 ± 4	28.3	0.18

[a] NA = Aluminium not listed as ingredient
SAS = Sodium aluminosilicate

table 6. Aluminium in salt

Brand [a]	Al contributing ingredient [b]	Al concentration (mg kg^{-1})	Approximate total packet or serving weight (g)	Al / serving (mg)
Single-serving Packet				
F01	SAS	190 ± 20	0.75	0.14
F02	SAS	180 ± 40*	0.75	0.13
F03	SAS	150 ± 45	1.5	0.22
F04	SAS	195 ± 28*	0.5	0.098
F05	NA	170 ± 45*	0.5	0.085
F06	NA	3 ± 2	0.5	0.0015
Multiple-serving container				
F07	SAS	125 ± 30	1.5	0.19
F08	NA	3 ± 1	1.5	0.0045

[a] F01, F02, F03, F05, F07, F08 made in USA, F04 and F06 made in Canada.
[b] NA = Aluminium not listed as ingredient
SAS = Sodium aluminosilicate
* Values based on 4 replicate observations.

table 7. Aluminium in pickle relish

Brand	**Product**	**Al contributing ingredient** [a]	**Al concentration (mg kg $^{-1}$)**	**Serving Size (g)**	**Al / serving (mg)**
G01	Sweet relish	Alum [b]	40 ± 5	15	0.6
G02	Sweet relish	NA	11 ± 1	15	0.2
G03	Dill relish	Alum	73 ± 40	15	1.1

[a] NA = Aluminium not listed as ingredient
[b] Alum = Al potassium sulfate

table 8. Quality control samples

Brand	Food Product (Matrix)	Form of Al added	Target Al Concentration (mg kg^{-1})	Determined Al concentration (mg kg^{-1}) [a]
C01	Classic sharp cheddar cheese	Kasal®	820	780 ± 20
E08	Non-dairy creamer	AlK_2SO_4	305	300 ± 30
F08	Salt	AlK_2SO_4	260	320 ± 20
D02	Baking mix	AlK_2SO_4	22000	21000 ± 400
D10	Cake mix	AlK_2SO_4	525	450 ± 40
D25	Pre-baked waffle, buttermilk	AlK_2SO_4	580	570 ± 50
G02	Sweet relish	Atomic absorption standard solution (Aldrich)	50	48 ± 4

[a] Results from five aliquots analyzed independently.